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**Disciplines**
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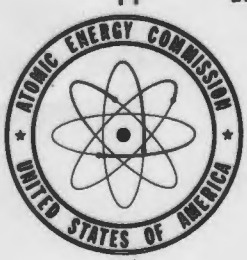


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# The Beta-Spectra of $P^{32}$ and $P^{33}$

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August 13, 1951

## Abstract

The beta-spectrum of $P^{32}$ has been examined with a thin-lens spectrometer. The maximum energy of the 14.3-day beta-activity, as determined from several Kurie plots, was found to be 1.704 ± 0.008 Mev. The Kurie plots gave excellent straight lines from the maximum beta-energy to about 0.26 Mev. In phosphorus samples prepared from neutron irradiated sulfur an additional beta-activity was observed having a maximum energy of 0.26 ± 0.02 Mev and a half-life of 24.8 ± 0.5 days. This low-energy beta-group was also observed in phosphorus samples prepared from sulfur and lithium chloride irradiated with X-rays having a maximum energy of 68 Mev. The low-energy beta-group was not observed in phosphorus samples prepared from sulfur irradiated with deuterons or phosphorus irradiated with neutrons. The low-energy beta-group is ascribed to $P^{33}$.

## I. Introduction

The beta-radiations from $P^{32}$ have been examined with various types of spectrometers by a number of investigators.[1]

[1] L. M. Langer and H. C. Price, Jr., Phys. Rev. 76, 641 (1949). Peacock, Jones, Overman, Plutonium Project Report Mon N-432, 56 (1947). K. Siegbahn, Phys. Rev. 70, 127 (1946); Nature 153, 221 (1944). C. M. Witcher, Phys. Rev. 60, 32 (1941). J. L. Lawson, Phys. Rev. 56, 131 (1939). E. M. Lyman, Phys. Rev. 51, 1 (1937).

Since the more recent of these investigations have shown that the Kurie plot of the $P^{32}$ spectrum is a good straight line, it was felt that this would be an excellent beta-spectrum with which to check the performance of a thin-lens spectrometer.[2]

[2] Jensen, Laslett, and Pratt, Phys. Rev. 75, 458 (1949).

The initial work on the beta-spectrum of $P^{32}$ was done in Dec. 1948. Since then the spectrometer has been modified to incorporate ring focusing.[3] In determining the beta-spectrum of

[3] Pratt, Boley, and Nichols, Rev. Sci. Instr. 22, 92 (1951). Keller, Koenigsberg, and Paskin, Rev. Sci. Instr. 21, 713 (1950).

$P^{32}$ a low-energy beta-component was observed in the Oak Ridge carrier-free samples which has a half-life of $24.8 \pm 0.5$ days and a maximum beta-energy of $0.26 \pm 0.02$ Mev. This half-life is appreciably greater than the 14.3 day half-life of the $P^{32}$ beta-component. Preliminary values of these results have been reported in the Progress Reports in Physics from the Ames

Laboratory at Iowa State College.[4]

[4]Ames Laboratory of the U. S. Atomic Energy Commission, Progress Reports in Physics, ISC-46, June 16, 1949; ISC-73, March 23, 1950; ISC-104, September 25, 1950; ISC-135, March 15, 1951. The results of a part of this work was reported at the Washington, D. C. meeting of the Am. Phys. Soc., April, 1951; Bull. Am. Phys. Soc. 26, No. 3, 24 (1951). At the same meeting Sheline, Holtzman, and Fan reported on a $P^{33}$ activity which was formed by gamma reactions on sulfur and chlorine; Bull. Am. Phys. Soc. 26, No. 3, 24 (1951).

During the progress of this investigation Agnew[5] and

[5]H. M. Agnew, Phys. Rev. 77, 655 (1950).

Warshaw, Chen, and Appleton[6] have reported on the beta-

[6]Warshaw, Chen, and Appleton, Phys. Rev. 80, 288 (1950).

spectrum of $P^{32}$ and observe a distinct excess of electrons in the momentum spectrum below about 300 kev. Agnew[5] states that "the $P^{32}$ was followed for five half-lives with no change in the Fermi plot." Warshaw et al.[6] state that "the spectrum has been followed for several half-lives with no change in shape." The data presented in this paper are in disagreement with these results since the half-life of 24.8 ± 0.5 days for the low-energy beta-group is appreciably different from the 14.3-day half-life of $P^{32}$. Our data indicate that there is an observable difference in shape of the beta-spectrum at low energies over a period of one half-life of $P^{32}$.

The low-energy beta-group present in the Oak Ridge carrier-free $P^{32}$ is believed to be due to $P^{33}$. This assignment is based primarily on the facts that this low-energy beta-group is not

present in phosphorus irradiated with neutrons or sulfur irradiated with deuterons, but is present in sulfur and lithium chloride irradiated with X-rays, from the Iowa State College Synchrotron, having a maximum energy of 68 Mev.

Recently Yaffe and Brown[7] have reported that they have

[7] L. Yaffe and F. Brown, Bull. Am. Phys. Soc. 26, No. 1, 45 (1951).

prepared $P^{33}$ by the reaction $P^{32}(n, \gamma)P^{33}$. They found a half-life of 22 ± 5 sec for this activity. Their results are based on the assumption that their target material was pure $P^{32}$. The data presented in this paper indicate that $P^{32}$ prepared by neutron irradiation of sulfur also contains initially about 2.5 atoms of $P^{33}$ per 100 atoms of $P^{32}$. Hence, in irradiating $P^{32}$ with neutrons it is possible to form both $P^{33}$ and $P^{34}$. We have found a half-life of 24.8 ± 0.5 days for $P^{33}$. Bleuler and Zünti[8] have reported a half-life of 12.4 sec for $P^{34}$. It

[8] E. Bleuler and W. Zünti, Helv. Phys. Acta 19, 137 (1946).

seems possible that the activity reported by Yaffe and Brown[7] is that of $P^{34}$ and not $P^{33}$.

All of the phosphorus samples examined with the thin-lens spectrometer were mounted on thin Formvar-polystyrene films having a surface density of about 40 $\mu$g/cm$^2$. The G-M counter window was a Formvar film of surface density about 0.3 mg/cm$^2$. No corrections have been made for the absorption due to the G-M counter window. The resolution of the spectrometer (full width at half maximum) was about 2.5 percent.

## II. Beta-Spectrum of $P^{32}$

The spectrum of the first sample[9] of carrier-free $P^{32}$, which was prepared at Oak Ridge about September 15, 1948, was determined as of December 23, 1948. This spectrum is shown in Fig. 1 where the data are plotted as obtained directly from the spectrometer, with the appropriate backgrounds subtracted.

Fig. 1 The Kurie plot of the spectrum shown in Fig. 1 gave a good straight line from the maximum energy down to about 0.26 Mev. Below this energy there was of course a definite excess of

Fig. 2 electrons. In Fig. 2 is shown a Kurie plot of a $P^{32}$ carrier-free source which had an average surface density of about 0.1 $mg/cm^2$. The beta-spectrum from which this Kurie plot was made

Fig. 3 is shown in Fig. 3 and is labeled 0 d. The Kurie plot is a good straight line and is not concave toward the energy axis as indicated by Agnew's[5] data. Several Kurie plots were made of beta-spectra obtained with various sources and all of them gave good straight lines from the maximum energy down to about 0.26 Mev. The maximum beta-energy shown in Fig. 2 represents an average value as determined from several Kurie plots. The average value of 1.704 ± 0.008 Mev is in good agreement with those determined by other investigators.[1,5,6]

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[9] We are indebted to Dr. C. W. Sherwin of the University of Illinois for this sample.

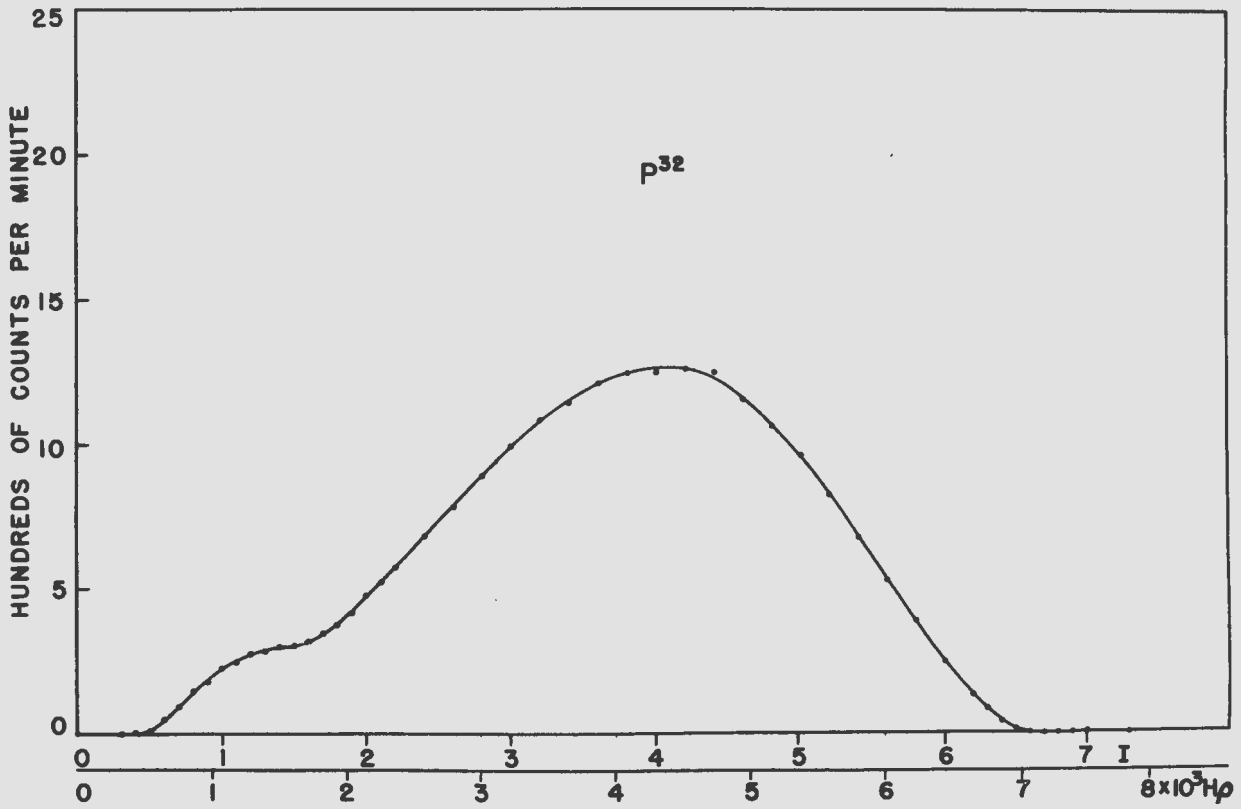


Fig. 1. Beta-spectrum of carrier-free $P^{32}$ which had decayed for about 99 days. A second beta-group is apparent at the low-energy end of the spectrum. I is the current in the spectrometer coil.

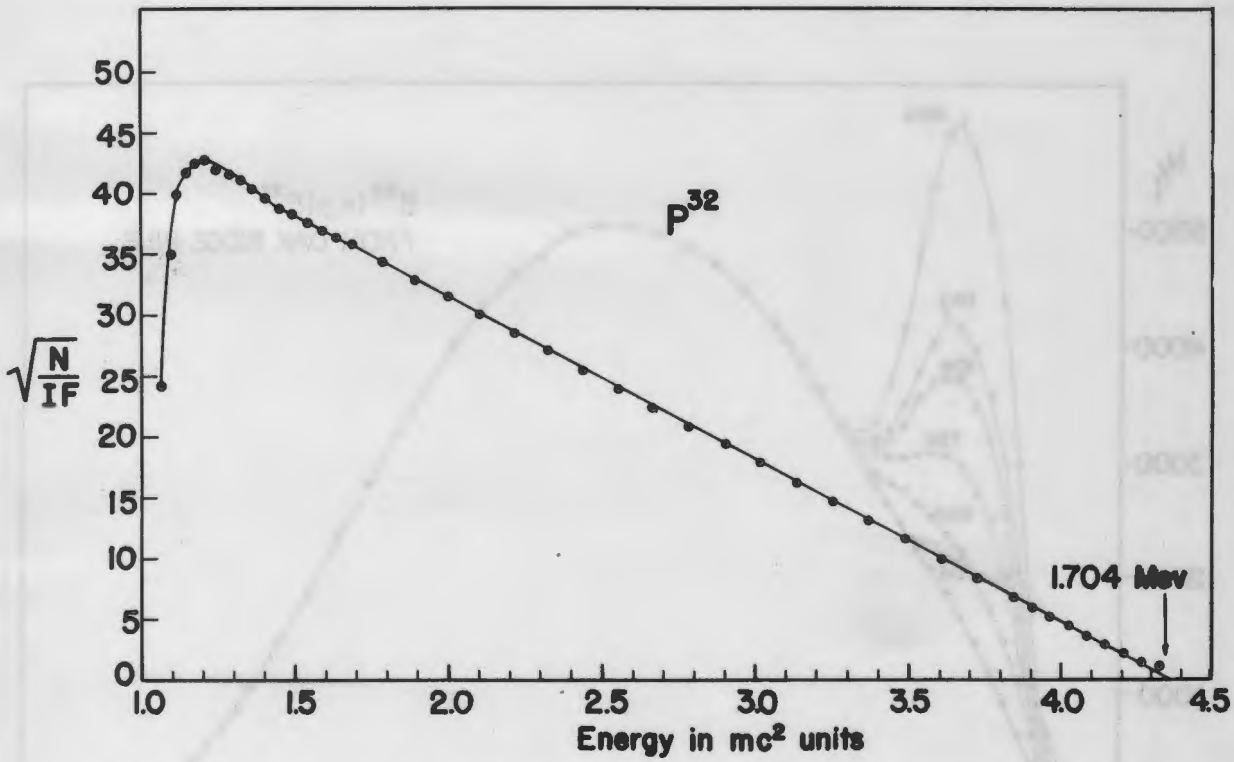


Fig. 2. Kurie plot of $P^{32}$ . The beta-spectrum from which this Kurie plot was made is shown in Fig. 3 and labeled 0 d. The maximum beta-energy of 1.704 ± 0.008 Mev represents the average value determined from several Kurie plots.

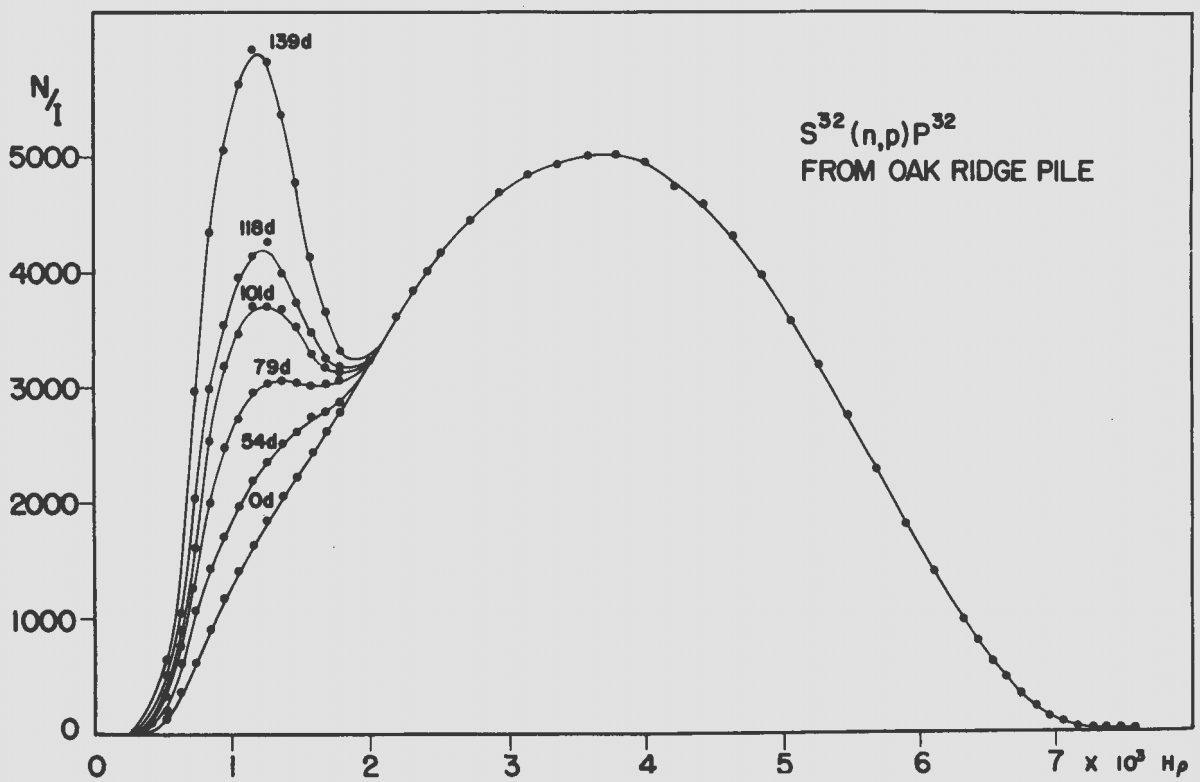


Fig. 3. Beta-spectra of a sample of carrier-free $P^{32}$ obtained at intervals over a period of 139 days. All curves have been normalized at the maximum counting rates of the main beta-group of $P^{32}$. The ordinate values are for the first spectrum, which is labeled 0 d. N is the number of counts per min and I is the current in the spectrometer coil.

## III. The Low-Energy Beta-Group

### A. Half-life and Maximum Beta-Energy

The spectrometer source from which the spectrum shown in Fig. 1 was obtained was followed for 36 days, during which time three additional beta-spectra were obtained. In each succeeding spectrum the low-energy beta-group became more prominent in comparison to the $P^{32}$ beta-component.

The second and third samples of carrier-free $P^{32}$ were received directly from Oak Ridge and their initial spectra determined as of July 29, 1949 and April 7, 1950 respectively. The second sample was followed with the spectrometer for 57 days, during which time four beta-spectra were determined. The third sample was followed for 139 days, during which time seven beta-spectra were determined. Six of these spectra are shown in Fig. 3 and have been normalized at the maximum counting rates of the $P^{32}$ beta-spectrum. The ordinates of the graph are applicable to the curve labeled 0 d. The first spectrum and the one determined 54 days later (labeled 0 d and 54 d respectively in Fig. 3) were obtained with one spectrometer source, while the other curves were obtained from a second and somewhat thicker spectrometer source, both of which were prepared from the third sample of carrier-free $P^{32}$. Samples one and two gave beta-spectra similar to those shown in Fig. 3. In the first beta-spectrum determined on samples two and three the low-energy beta-group could not be observed with certainty and could readily be attributed to the effect of a thick source.

The half-life of the low-energy beta-group was determined from the spectra shown in Fig. 3 by determining the ratios of

the areas of the low-energy beta-group to that of the $P^{32}$ as a function of time. The natural logarithms of these ratios

Fig. 4 are plotted as a function of time in Fig. 4. The time at which the first beta-spectrum was determined is taken as the time reference. The slope of the straight line gives the difference in the decay constants of the two activities. Using 14.3 days as the half-life of $P^{32}$ the data shown in Fig. 4 gives a half-life of 24.2 days for the low-energy beta-group. The activity of a portion of the third sample of carrier-free $P^{32}$ was followed by means of a G-M counter for a period of 25 days after it had decayed for 10 months. At this time the activity of the low-energy group was estimated to be about 90 percent

Fig. 5 of the total activity. These data are shown in Fig. 5. The half-life, as determined by the method of least squares, was found to be 25.2 days. Another portion of the same sample of carrier-free $P^{32}$ has been followed with a G-M counter for a period of 82 days after it had decayed for 404 days. These data reveal the presence of an additional weak activity having a half-life somewhat longer than 25 days.

From Fig. 4 it is seen that the 24.8-day activity, which we ascribe to P33, was about 1.4 percent of the $P^{32}$ activity at the time the first beta-spectrum of the sample was determined. The ratio of these activities was somewhat smaller at the time the sample was removed from the pile, but unfortunately this time is not know. No correction has been made for the absorption due to the G-M counter window. This correction would increase the ratio of the activities.

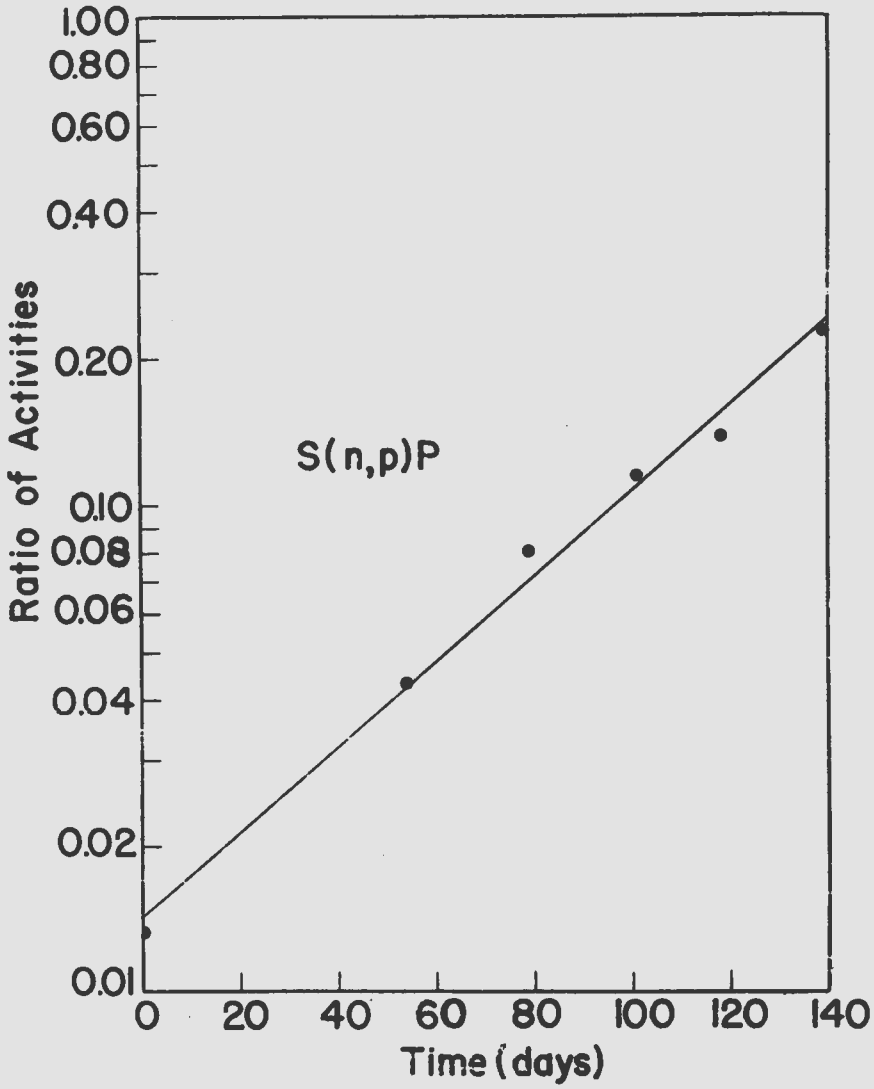


Fig. 4. A plot of the ratios of the areas of the low-energy beta-group to that of $P^{32}$ as a function of time as given by the curves of Fig. 3. The time at which the first beta-spectrum was determined is taken as the time reference.

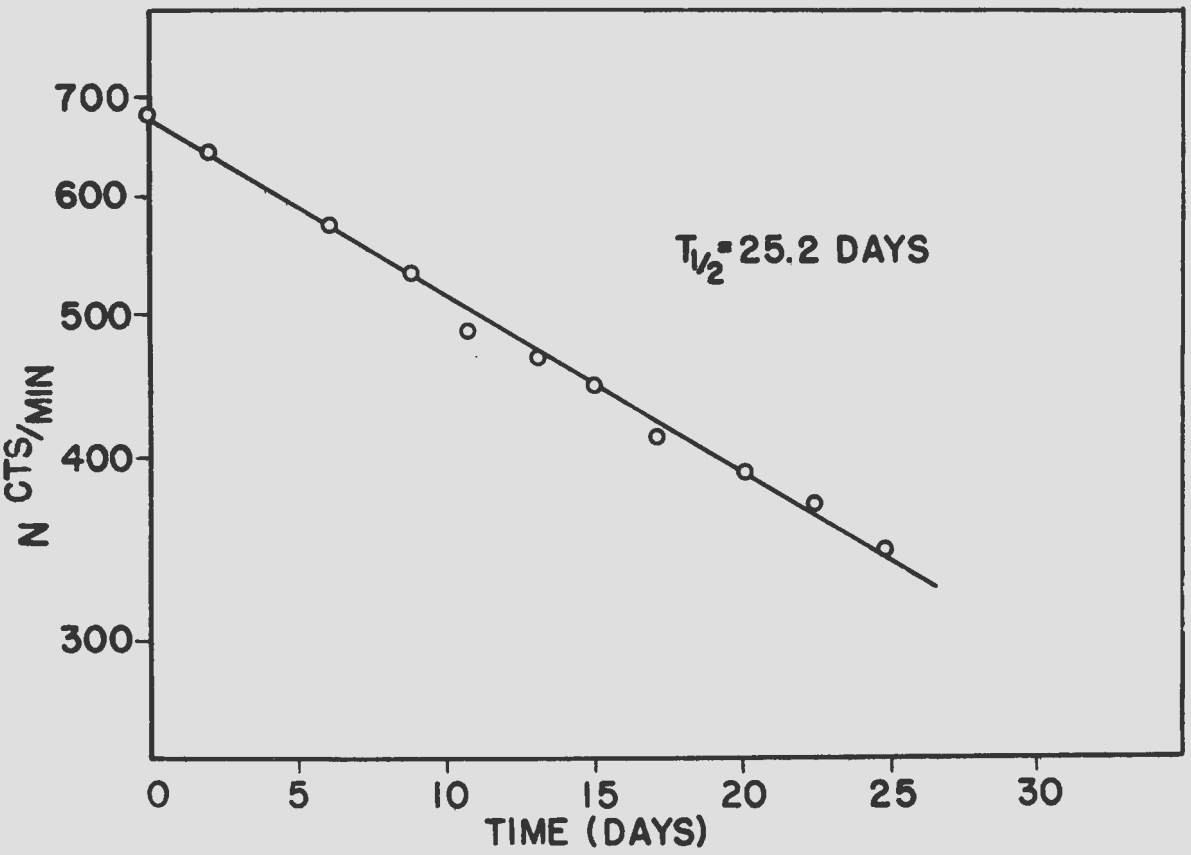


Fig. 5. Decay curve of low-energy beta-group in $S^{32}(n,p)$ $P^{32}$ sample. The sample had decayed for 10 months before these data were obtained. It is estimated that the activity of the low-energy beta-group was about 90 per cent of the total activity.

The maximum energy of the low-energy beta-group, which was determined from the various beta-spectra and Kurie plots by noting the energy at which the low-energy beta-group became apparent on the respective curves, was found to be 0.26 ± 0.02 Mev. An effort was made to obtain Kurie plots of the low-energy beta-group, but these were unsatisfactory due to the thick sources used.

By deflection of the beta-particles in a magnetic field it was found that the low-energy beta-group consists of negative electrons. Cloud chamber pictures obtained with a sample of $P^{32}$ that had decayed for 12 months, at which time it was estimated that the activity of the low-energy group was about 96 percent of the total activity, also confirmed the fact that the radiation consists of negative electrons.

There is a possibility that the low-energy beta-group could be due to a contaminant that is not sufficiently removed by the chemical purification process of the neutron irradiated sulfur. The first consideration is that of $S^{35}$. However, the half-life and maximum beta-energy of $S^{35}$ are 87.1 days and 0.166 Mev[10] respectively. This possibility is excluded since

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[10]Way, Fano, Scott, and Thew, Nuclear Data, Nat. Bur. Standards Circular 499 (1950).

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these values are not in agreement with those of the low-energy beta-group given above. $S^{37}$ is not a possibility since its half-life is only 5.0 min.[10] In fact there is no known beta-activity with a half-life of 24.8 ± 0.5 days and maximum beta-energy of 0.26 ± 0.02 Mev.

If the low-energy beta-group is due to a contaminant, the amount of it present in waste products separated from the $P^{32}$ during chemical purification would perhaps be considerably greater than in the carrier-free $P^{32}$. One such waste fraction was obtained from Oak Ridge and its spectrum determined with the spectrometer. This sample was rather weak, but practically all of the activity was due to $P^{32}$. The low-energy group was present, but not to any greater extent than in other carrier-free $P^{32}$ samples. Hence, the low-energy beta-group does not appear to be due to a contaminant in the target material.

The carrier-free $P^{32}$ produced at Oak Ridge is formed by a $S^{32}(n,p)P^{32}$ reaction. Since the isotopes of sulfur are 32 (95.00%), 33(0.74%), 34(4.24%), and 36(0.017%)[11] the nuclear

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[11] Way, Wood, and Thew, Nuclear Data, Supplement 1, Nat. Bur. Standards Circular 499 (1951).

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reactions $S^{33}(n,p)P^{33}$, $S^{34}(n,p)P^{34}$, and $S^{36}(n,p)P^{36}$ would also be possible. $P^{34}$ is known to have a half-life of 12.4 sec[8] and is therefore eliminated as a possibility. This leaves the possibility of the low-energy beta-group as being due to $P^{33}$ or $P^{36}$. Since $S^{36}$ has a very low abundance the cross section for the $S^{36}(n,p)P^{36}$ reaction would have to be very large if the low-energy beta-group were due to $P^{36}$. This is, of course, a possibility. Another possibility is that $P^{33}$ is formed by a second order $(n,\gamma)$ reaction on $P^{32}$. This is very unlikely unless $P^{32}$ has an extremely large thermal neutron cross section. For a 28 days irradiation of sulfur at a flux of $5 \times 10^{11}$

neutrons/$cm^2$/sec the thermal neutron cross section of $P^{32}$ would have to be about 31,000 barns in order for the activity of the $P^{33}$ to be one percent of the $P^{32}$ activity at the end of the irradiation time.

B. Beta-Spectra of $P^{32}$ Prepared by Methods other than S(n,p)P

Radioactive $P^{32}$ can also be prepared by a $P^{31}(n,\gamma)P^{32}$ reaction.[12] Since stable phosphorus consists of a single isotope,[13] $P^{32}$ is the only isotope formed by a $(n,\gamma)$ reaction. It would be possible to form $P^{33}$ by a second order $(n,\gamma)$ reaction on $P^{32}$. If $P^{33}$ were to be detected in the spectrometer by this reaction the cross-section for the $P^{32}(n,\gamma)P^{33}$ reaction would have to be very large. For a 14 day irradiation of $P^{31}$ at a flux of $3 \times 10^{13}$ neutrons/$cm^2$/sec the thermal neutron cross section of $P^{32}$ would have to be about 980 barns in order for the activity of the $P^{33}$ to be one percent of the $P^{32}$ activity at the end of the irradiation time. The thermal neutron cross section of $P^{31}$ is 0.15 barn.[14]

A sample of $P^{32}$, prepared by a $P^{31}(n,\gamma)P^{32}$ reaction, was obtained from Oak Ridge. As reported previously[15] this sample

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[12] Seren, Friedlander, and Turkel, Phys. Rev. 72, 888 (1947).

[13] F. W. Aston, Mass Spectra and Isotopes (E. Arnold and Co., London, 1942).

[14] H. Pomerance, ORNL-577, 25 (1949) and ORNL-366, 43 (1949).

[15] Erling N. Jensen and R. T. Nichols, Bull. Am. Phys. Soc. 26, No. 3, 24 (1951).

appeared to have a low-energy beta-group similar to the one observed in the $P^{32}$ samples prepared by neutron irradiation of sulfur. However, the data were not conclusive since the sample had a low specific activity and therefore could not be followed in the spectrometer for a sufficiently long period of time. Two other higher specific activity sources of $P^{32}$, prepared by a $P^{31}(n, \gamma)P^{32}$ reaction, were obtained, one from Oak Ridge and one from Chalk River. The low-energy beta-group was not observed in either one of these samples. The appearance of the low-energy beta-group in the first $P^{32}$ sample prepared by a $P^{31}(n, \gamma)P^{32}$ reaction is unexplained.

The Chalk River $P^{32}$ sample was prepared by irradiating red phosphorus for 14 days with a total thermal neutron exposure of $3.65 \times 10^{19}$ neutrons/$cm^2$,[16] and was removed from the pile on

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[16] K. R. Manning, Chalk River, Ont., private communication.

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March 5, 1951. The beta-ray spectra of this sample are shown in Fig. 6, and again are normalized at the maximum counting rates. The ordinates of the graph apply to the data obtained as of March 20, 1951. Since it was of interest to follow the phosphorus sample in the spectrometer for as long a time as possible, the spectrometer source was made quite thick and this accounts for the distortion of the spectra at the lower energies as shown by a spectrum obtained from a thin source of the same sample. The sample was followed with the spectrometer for 112 days during which time four spectra were obtained. The first and last spectra shown in Fig. 6 were obtained 15 days and 127 days respectively after the sample was removed

Fig. 6

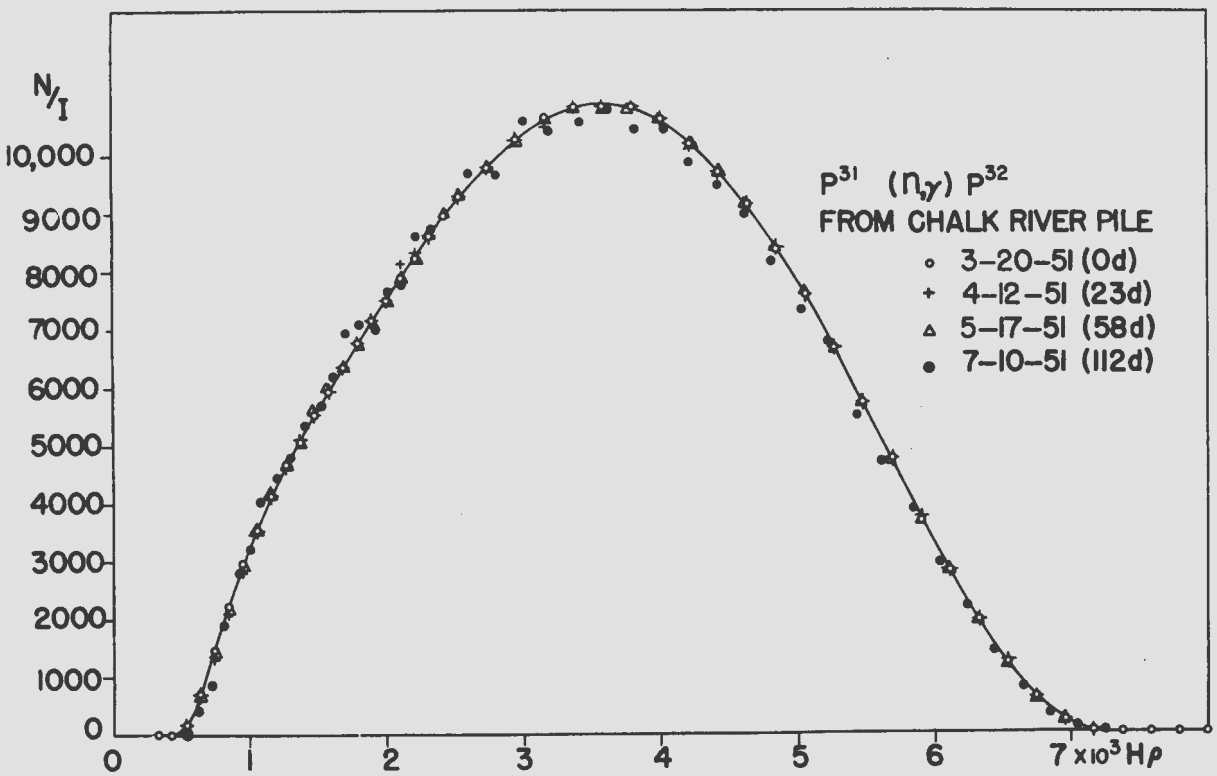


Fig. 6. Beta-ray spectra of $P^{32}$ sample prepared by a $P^{31}(n,\gamma)P^{32}$ reaction. These four spectra were obtained over a period of 112 days. The spectra have been normalized at the maximum counting rates. The ordinates apply to the spectrum determined as of March 20, 1951. The distortion of the spectra at low energies is due to a thick source. The last spectrum was determined 127 days after the sample was removed from the pile. N is the number of counts per min and I is the current in the spectrometer coil.

from the Chalk River pile. As can be seen from Fig. 6 all four spectra are essentially identical and there is no evidence of the low-energy beta-group observed in the Oak Ridge carrier-free $P^{32}$ prepared by neutron irradiation of sulfur. The spectrum shown in Fig. 6 which was determined as of July 10, 1951 had a maximum N/I value of 43.8.

The second sample of $P^{32}$ prepared by a $P^{31}(n, \gamma)P^{32}$ reaction, which was received from Oak Ridge, was followed with the spectrometer for a period of 47 days during which time three spectra were obtained. The first and last spectra of this sample were obtained 45 days and 92 days respectively after the sample was removed from the pile. As in the case of the Chalk River sample the three spectra were essentially identical and there was no evidence of the low-energy beta-group. It is therefore concluded that the low-energy beta-group observed in the Oak Ridge carrier-free $P^{32}$, which is prepared by neutron irradiation of sulfur, is not present in $P^{32}$ samples prepared by a $P^{31}(n, \gamma)P^{32}$ reaction.

Since the low-energy beta-group is present in $P^{32}$ samples prepared by neutron irradiation of sulfur but is not present in $P^{32}$ samples prepared by neutron irradiation of phosphorus it follows that the low-energy beta-group cannot be due to a second order $(n, \gamma)$ reaction on $P^{32}$. If this were the case the low-energy beta-group would be much more prominent in the Chalk River sample than in the Oak Ridge $P^{32}$ samples prepared by neutron irradiation of sulfur due to the higher neutron flux of the Chalk River pile. However, it does not rule out the possibility that the low-energy beta-group is due to $P^{33}$

or possibly $P^{36}$ as a result of (n,p) reactions on $S^{33}$ and $S^{36}$ respectively.

The isotope $P^{32}$ can also be prepared by a $S^{34}(d,\alpha)P^{32}$ reaction.[17] A sulfur sample was irradiated for two hours with a 16 Mev deuteron beam of approximately 15 $\mu$amp [18] at the University of Pittsburgh cyclotron. After receiving the irradiated sulfur a chemical separation of phosphorus was performed on this sample. A description of the chemical procedure is given in Appendix A. The phosphorus sample was then followed in the spectrometer for a period of 55 days during which time three beta-spectra were obtained. These spectra are shown in Fig. 7 and are normalized at the maximum counting rates. The spectrometer source was quite thick and this again accounts for the distortion at the lower energies. The sulfur sample was irradiated with deuterons on Jan. 28, 1951. Hence, the first and last spectra shown in Fig. 7 were obtained 16 days and 71 days respectively after the deuteron bombardment. The ordinates of Fig. 7 apply to the data obtained as of Feb. 13, 1951. From Fig. 7 it is seen that the three spectra are essentially identical and that there is no evidence of the low-energy beta-group observed in the Oak Ridge carrier-free samples of $P^{32}$ prepared by neutron irradiation of sulfur.

Fig. 7

A (d,$\alpha$) reaction on all the isotopes of sulfur would form $P^{30}$, $P^{31}$, $P^{32}$, and $P^{34}$. Since neither $P^{33}$ or $P^{36}$ is formed by

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[17] R. Sagane, Phys. Rev. 50, 1141 (1936).

[18] A. J. Allen, private communication.

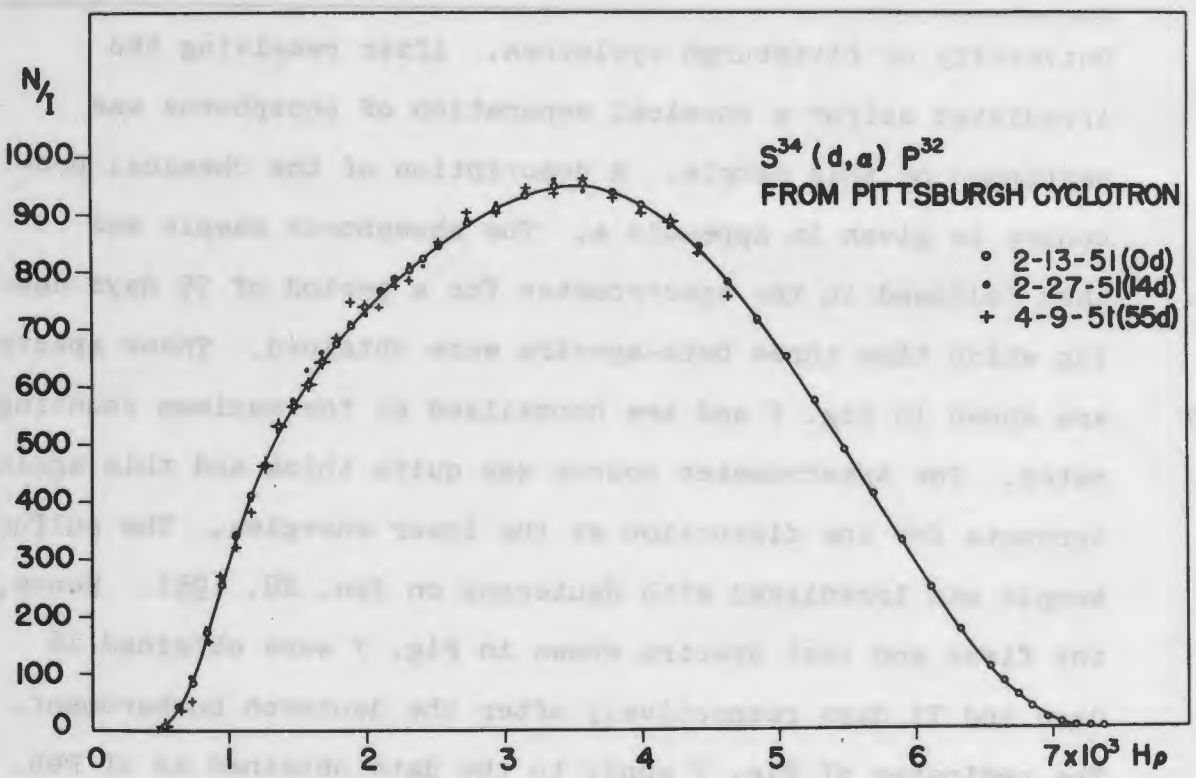


Fig. 7. Beta-spectra of $P^{32}$ sample prepared by a $S^{34}$ (d,$\alpha$)$P^{32}$ reaction. These three spectra were obtained over a period of 55 days. The spectra have been normalized at the maximum counting rates. The ordinates apply to the spectrum determined as of Feb. 13, 1951. The distortion of the spectra at low energies is due to a thick source. The last spectrum was determined 71 days after the deuteron bombardment. N is the number of counts per min. and I is the current in the spectrometer coil.

this reaction and since $P^{32}$ is the only activity apparent in Fig. 7 it follows that these data are also consistent with the possibility that the low-energy beta-group observed in $P^{32}$ samples prepared by neutron irradiation of sulfur is due to $P^{33}$ or possibly $P^{36}$.

Another method of forming $P^{32}$ is by a ($\gamma$,p) reaction on sulfur. This reaction on all the isotopes of sulfur would form $P^{31}$, $P^{32}$, $P^{33}$ and $P^{35}$. Perlman[19] has reported that at 50 Mev and 100 Mev the ($\gamma$,pn) yield on the reaction $Ge^{70}$($\gamma$,pn)$Ga^{68}$ is of the same order as ($\gamma$,p) yields on five isotopes reported by Perlman and Friedlander.[20] Other ($\gamma$,pn) reactions have been reported by Hoffman[21] [$A^{40}$($\gamma$,pn)$Cl^{38}$], Moses[22] [$W^{182}$($\gamma$,pn)$Ta^{180}$], and Katz and Penfold[23] [$S^{32}$($\gamma$,pn)$P^{30}$]. Moses[22] also found that the yield for the ($\gamma$,pn) reaction on $W^{182}$ was of the same order as the ($\gamma$,p) reaction on W at maximum X-ray energies of 30 Mev and 68 Mev. Hence, in irradiating sulfur with X-rays one would expect to form $P^{30}$, $P^{31}$, $P^{32}$, and $P^{34}$ by ($\gamma$,pn) reactions. The radioisotopes expected to be formed by irradiating sulfur with X-rays, and having appreciable half-lives, are those of $P^{32}$ and $P^{33}$.

[19] M. L. Perlman, Phys. Rev. 75, 988 (1949).

[20] M. L. Perlman and G. Friedlander, Phys. Rev. 74, 442 (1948).

[21] M. Hoffman, Bull. Am. Phys. Soc. 26, No. 3, 24 (1951).

[22] A. J. Moses, Masters Thesis, Iowa State College (1950).

[23] L. Katz and A. S. Penfold, Phys. Rev. 81, 815 (1951).

Sulfur was irradiated for about 30 hours with X-rays, from the Iowa State College synchrotron, that had a maximum energy

of 68 Mev. A phosphorus separation was performed on the irradiated sulfur. The chemical procedure followed in this phosphorus separation is given in Appendix B.

The synchrotron-produced phosphorus sample was followed by means of a G-M counter having a window surface density of 2.3 mg/cm$^2$. The data obtained are shown in Fig. 8. The solid circles represent the total radiation from the sample. The crosses represent the corrected counting rates obtained with an aluminum absorber between the sample and the counter. The absorber had a surface density of 75 mg/cm$^2$, which is sufficient to absorb completely electrons having an energy of 0.29 Mev.[24]

Fig. 8

[24] L. E. Glendenin, Nucleonics 2, No. 1, 12 (1948).

This completely absorbed the low-energy beta-group and therefore only the betas from $P^{32}$ were counted. In order to obtain the total counting rate due to the $P^{32}$ activity the counting rates of a $P^{32}$ sample, prepared by a $P^{31}(n,\gamma)P^{32}$ reaction, were obtained with and without the same absorber and for the same geometry. The ratio of these two counting rates gave the correction factor by which the counting rates of the synchrotron produced activity, with absorber, were multiplied in order to obtain the total counting rate due to the $P^{32}$ activity. By the method of least squares the half-life of the activity shown by the crosses in Fig. 8 was found to be 14.3 days, which is in good agreement with the published values of the half-life[10] of $P^{32}$. It is clear from these two sets of data that there is more than one activity present in the synchrotron-produced phosphorus. By subtracting from the total activity the counting rates due to the $P^{32}$ activity the open circles of Fig. 8 are obtained. This indicates that there are

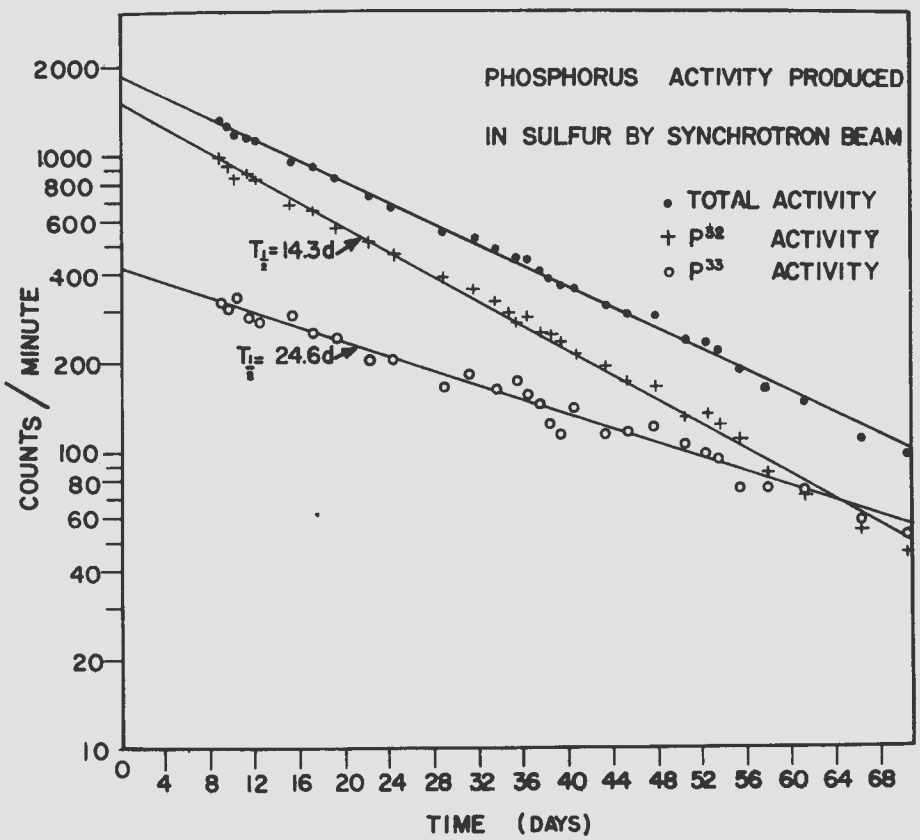


Fig. 8. Phosphorus activities produced in sulfur irradiated with X-rays from the synchrotron. The solid circles represent the total activity while the crosses represent the corrected counting rates obtained with a 75 mg/cm$^2$ aluminum absorber. The open circles, which were obtained by subtracting the $P^{32}$ activity from the total activity, represent the activity of the low-energy beta-group.

two activities present in the synchrotron-produced phosphorus and that the half-life of the second activity is 24.6 days. This is in good agreement with the half-life values of 25.2 and 24.2 days found for the low-energy beta-groups in the carrier-free $P^{32}$ prepared by neutron irradiation of sulfur. A previous run on sulfur irradiated with X-rays from the synchrotron gave half-life values of 14.4 days for the $P^{32}$ activity and 25.2 days for the low-energy beta-group.

After corrections were applied for the counter window, air absorption, cellophane covering of the source and self scattering due to source weight[25] to the initial activities of

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[25]Engelkemeir, Seiler, Steinberg, and Winsberg, National Nuclear Energy Series 9, paper 4, 56 (1951).

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$P^{32}$ and the low-energy beta-group as given by Fig. 8 it was found that the initial activity of the low-energy beta-group was 45 percent of the total activity. This is about the activity expected for a 30 hr irradiation of sulfur on the assumption that the cross sections for ($\gamma$,p) on $S^{33}$ and $S^{34}$ and ($\gamma$,np) on $S^{34}$ are all equal. This activity is much larger than the 1.4 percent found in the phosphorus samples prepared by neutron irradiation of sulfur. This is to be expected since the abundances of $S^{33}$ and $S^{34}$ are 0.74 percent and 4.24 percent respectively.

A sample of lithium chloride was also irradiated with the X-rays from the synchrotron at the same time as the sulfur sample was irradiated. The phosphorus separation performed on this sample is given in Appendix B. This phosphorus sample

was followed by means of the same G-M counter and in precisely the same manner as the phosphorus sample obtained from the synchrotron irradiation of sulfur. These data are shown in Fig. 9. In this case the half-life of the $P^{32}$ activity was determined as 15.2 days while the low-energy beta-group was found to have a half-life of 24.7 days which is in excellent agreement with that determined from Fig. 8. The phosphorus activities are presumably due to the reactions $Cl^{35}(\gamma,2p)P^{33}$, $Cl^{35}(\gamma,n2p)P^{32}$, and $Cl^{37}(\gamma,\alpha)P^{33}$. Again applying the appropriate corrections it was found that the initial activity of the low-energy beta-group was 48 percent of the total activity.

Fig. 9

C. Discussion

In the experiments described above radioactive phosphorus has been produced by five different reactions. In all of these reactions, except $P^{31}(n,\gamma)P^{32}$, a phosphorus separation was made on the irradiated material before the radiations were examined. Table I gives a summary of the results and the possible isotopes formed by each reaction. The isotopes $P^{30}$ and $P^{34}$ can be eliminated from consideration since their half-lives are 2.18 min[26] and 12.4 sec[8] respectively. The only other

[26] J. Cichocki and A. Soltan, Compt. rend. 207, 423 (1938).

isotope, in addition to $P^{32}$, that is present in all three reactions $\left[S(n,p)P,\ S(\gamma,pn)P,\text{ and } Cl\left(\begin{matrix}\gamma,2p\\ \gamma,n2p\\ \gamma,\alpha\end{matrix}\right)P\right]$ in which the low-energy beta-group is present is that of $P^{33}$. Neither $P^{35}$ nor $P^{36}$ is formed in all three of these reactions. It is therefore

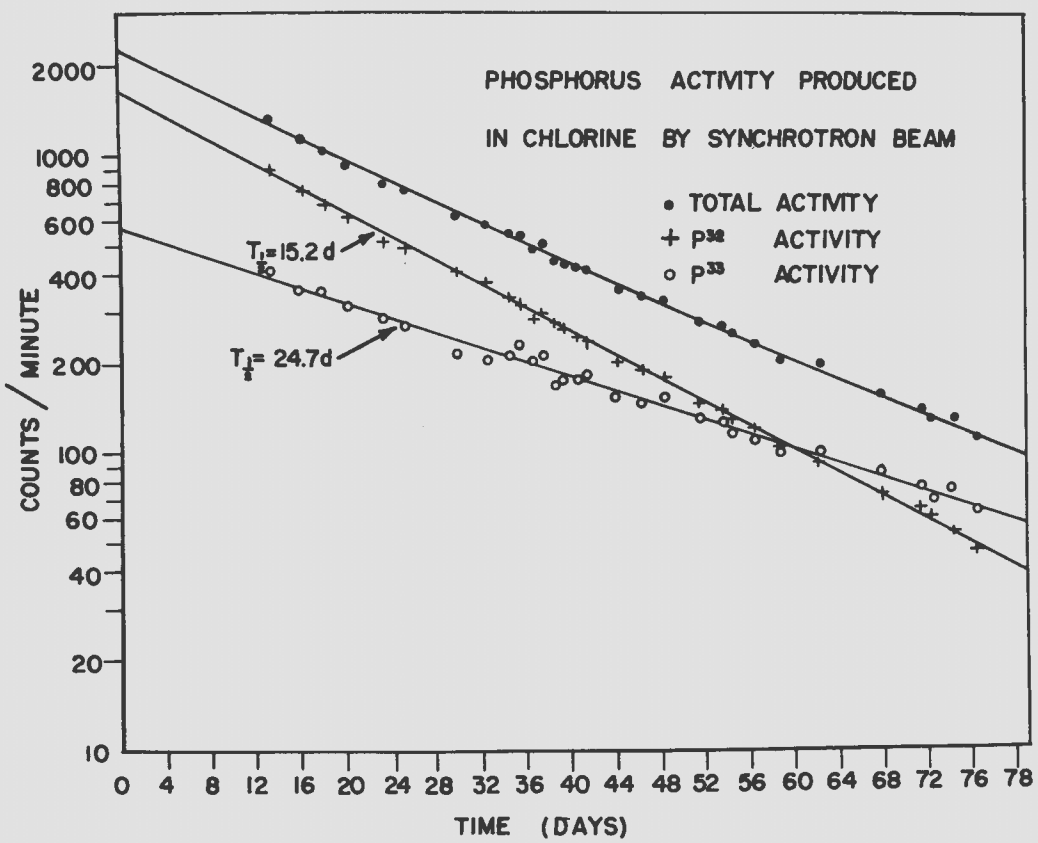


Fig. 9. Phosphorus activities produced in lithium chloride irradiated with X-rays from the synchrotron. The solid circles represent the total activity while the crosses represent the corrected counting rates obtained with a 75 mg/cm$^2$ aluminum absorber. The open circles, which were obtained by subtracting the $P^{32}$ activity from the total activity, represent the activity of the low-energy beta-group.

Table I

Summary of results on phosphorus activities

produced by various reactions

| Reaction | Presence of $P^{32}$ | Presence of low-energy beta-group | Possible formation of phosphorus isotopes |
|---|---|---|---|
| 1. S(n,p)P | Yes | Yes | 32, 33, 34, 36 |
| 2. $P^{31}(n,\gamma)P^{32}$ | Yes | No | 32 |
| 3. S(d,$\alpha$)P | Yes | No | 30, 31, 32, 34 |
| 4. S($\gamma$,p / $\gamma$,pn)P | Yes | Yes | 30, 31, 32, 33, 34, 35 |
| 5. Cl($\gamma$,2p / $\gamma$,n2p / $\gamma$,$\alpha$)P | Yes | Yes | 31, 32, 33, 34, 35 |

concluded that the low-energy beta-group observed in the carrier-free $P^{32}$ prepared at Oak Ridge, by neutron irradiation of S, is due to $P^{33}$. The $P^{33}$ is formed by the reaction $S^{33}(n,p)P^{33}$. Our results indicate that $P^{33}$ decays by negatron emission with a maximum beta-energy of 0.26 ± 0.02 Mev and a half-life of 24.8 ± 0.5 days. This half-life value is the mean of the five experimental values. The ratio of the initial activity of $P^{33}$ to $P^{32}$ in carrier-free $P^{32}$ is about 1.4 percent.

Using a maximum beta-energy of 0.26 Mev, a half-life of 24.8 days and an f value of 0.055, obtained from the graphs by Feenberg and Trigg,[27] it is found that $P^{33}$ has a log ft value of 5.1. According to Nordheim[28] this is a normal allowed transition. The spin of $S^{33}$ is known to be 3/2.[29,30] The magnetic moment[30] of $S^{33}$ indicates a $d_{3/2}$ state. According to the nuclear shell model,[31] a regular filling of the levels would give a $d_{3/2}$ state to the $P^{33}$ nucleus. The decay of $P^{33}$ would then be a normal allowed transition in which the orbital momentum is unchanged and with a spin change of zero. For

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[27] E. Feenberg and G. Trigg, Revs. Modern Phys. 22, 399 (1950).

[28] L. W. Nordheim, Phys. Rev. 78, 294 (1950).

[29] C. H. Townes and S. Geschwind, Phys. Rev. 74, 626 (1948).

[30] Eshbach, Hillger, and Jen, Phys. Rev. 80, 1106 (1950).

[31] M. G. Mayer, Phys. Rev. 78, 16 (1950).

such a transition the values of log ft range mostly from 4.8 to 5.5.[28] The experimental value of 5.1 for log ft is in good agreement with these values. However, $P^{31}$ is known to have a spin of 1/2 with a $S_{1/2}$ state. On the basis of the nuclear shell model one would expect $P^{33}$ to have a $S_{1/2}$ state also. The decay of $P^{33}$ would then involve a transition in which the orbital momentum changes by two units with a spin change of one unit. Transitions of this type have log ft values[28] mostly in the range 6.5 to 7.5, but with some stragglers. The experimental value of 5.1 for log ft is not in agreement with these values. These considerations add confirmation to the $d_{3/2}$ state of $P^{33}$.

During the early part of the work on the beta-spectrum of $P^{32}$, when it erroneously appeared that the low-energy beta-group was present in $P^{31}$ irradiated with neutrons, it was thought that perhaps the low-energy beta-group was due to a daughter product of $P^{32}$. At this time Dr. L. G. Elliott of Chalk River, Ont. suggested the possibility of simultaneous emission of two electrons or of two quanta between two states having zero spin and opposite parity. Koenigsberg and Keller[32]

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[32]E. Koenigsberg and J. M. Keller, private communication. These calculations were made following the theory of R. G. Sachs, Phys. Rev. 57, 194 (1940).

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have calculated that for the case under consideration here the transition probability of two quanta emission is about 2.2 x $10^3$ times as large as for two electron emission. Also no net coincidence counts were observed, as would be expected for

simultaneous emission of two electrons or two quanta, in a $P^{32}$ sample that had decayed for 257 days. To check on the possibility of an isomeric state in sulfur a large batch of carrier-free $P^{32}$ was allowed to decay for 111 days at Oak Ridge. Samples from this batch were obtained when it was first set aside and then 111 days later after the batch had been put through a second chemical purification process which presumably removed any sulfur present. This should have altered the ratio of the activity of the low-energy beta-group, to that of $P^{32}$ if there is an isomeric state in sulfur. From the beta-spectra of these samples it was found that the ratio of the activities was not changed due to the second chemical purification. The nuclear shell model[31] indicates that one would not expect to find an isomeric state in sulfur. The fact that the low-energy beta-group is not observed in all of the reactions in which $P^{32}$ was observed, even though the samples were followed for many half-lives, eliminates the possibility of the low-energy beta-group as being due to a daughter product of $P^{32}$.

The results of this paper in which the low-energy beta-group is ascribed to $P^{33}$ with a maximum beta-energy of 0.26 ± 0.02 Mev and a half-life of 24.8 ± 0.5 days are in disagreement with those reported by Yaffe and Brown[7] in which they give a half-life of 22 ± 5 seconds for $P^{33}$. Their assignment of $P^{33}$ is based on the assumption that their target material was pure $P^{32}$. Our results indicate that $P^{32}$ prepared by neutron irradiation of sulfur contains about 2.5 atoms of $P^{33}$ per hundred

atoms of $P^{32}$. It is possible that the activity measured by Yaffe and Brown was that of $P^{34}$ produced by a $P^{33}(n, \checkmark)P^{34}$ reaction. The $P^{33}$ would not be observed unless the activity was followed for several half-lives of $P^{32}$.

Sheline, Holtzman and Fan[33] have recently reported observ-

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[33] Sheline, Holtzman, and Fan, Phys. Rev. ____, (  ).

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ing a low-energy beta-group in sulfur and chlorine irradiated with the 48 Mev X-ray spectrum of the University of Chicago Betatron and also in old Oak Ridge carrier-free $P^{32}$ samples. They also assign the low-energy beta-group to $P^{33}$ and give a maximum beta-energy of 0.27 ± 0.02 Mev and a half-life of 25 ± 2 days. Our results are in good agreement with these.

## IV. Acknowledgments

The authors wish to express their appreciation to Dr. L. J. Laslett for many helpful suggestions and his assistance in obtaining part of the data, particularly the determination of the charge of the $P^{33}$ radiation; to Dr. A. F. Voigt and Messrs. L. McIsaac and A. Richardson for the chemical separation performed on the deuteron bombardment of sulfur and preparation of spectrometer sources; to Dr. D. S. Martin, Jr. and Messrs. F. J. Hughes and W. R. Daniels for the chemical separations performed on the synchrotron bombardments of sulfur and lithium chloride and the obtaining of the data on these phosphorus samples; to Dr. D. J. Zaffarano and the synchrotron crew for the X-ray irradiation of sulfur and lithium

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A new electronic current control for the thin-lens spectrometer has been designed, constructed and put into operation by Mr. A. Read and for this the authors wish to express their appreciation.

## Appendix A

The following phosphorus separation[34] was performed on the sulfur irradiated with 16 Mev deuterons. The irradiated

[34] The chemical procedure for this phosphorus separation was received, by private communication, from J. H. Gillette, superintendent Radioisotope Control Department, Oak Ridge National Laboratory.

sulfur (approximately two grams) was melted in a pyrex beaker and poured into another beaker containing 15 ml of boiling fuming nitric acid. The mixture was boiled for one hour, after which time the sulfur had settled to the bottom. The supernatant liquid was poured off and boiled again to oxidize any colloidal sulfur and also to remove excess $NO_2$. When this solution became clear, two mg of $La(NO_3)_3$ were added. This was followed by a careful neutralization with $NH_4OH$ until the precipitation of $La(OH)_3$ was complete. The precipitate was filtered on a medium sintered glass disk and washed with dilute $NH_4OH$. Following the washing, the precipitate was dissolved with 10 ml of hot 6 N HCl and washed with sufficient water to give a final concentration of 0.1 N in HCl. This solution was passed through a Dowex-50 column to replace the metal cations by hydrogen ions. To the effluent one mg of P, as $H_3PO_4$, was added and the solution evaporated to dryness at 90 - 100°C. The residue was dissolved in a minimum amount of water and filtered to remove any resin solids. The filtrate was evaporated to near dryness and mounted on a Formvar-polystyrene film.

## Appendix B

The separation of phosphorus from sulfur irradiated with X-rays from the Iowa State College synchrotron was essentially a boiling nitric acid extraction described by Cohn[35] and

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[35]W. E. Cohn, Clinton Laboratories, MDDC-518 (1946).

studied by Kenny and Spragg.[36] In addition to this separation

[36] A. W. Kenny and W. T. Spragg, AERE-C/R-485 (1950).

it was desirable to remove any trace amounts of other radioisotopes which might have been formed. To accomplish this 10 mg each of cupric, chromic, arsenate and phosphate ions were added as carriers. The solution was diluted with 3 N HCl and saturated with $H_2S$. After standing for several hours the copper and arsenic sulfides were removed by centrifuging. The supernatant liquid was boiled in order to remove the excess $H_2S$. HCl was then removed by boiling with $H_2SO_4$ and the solution was neutralized with $NH_3$. The chromium was oxidized to chromate with $Na_2O_2$. The solution was acidified with HCl and $FeCl_3$ solution was added. An excess of $NH_3$ was added in order to precipitate hydrated $Fe(OH)_3$. This was dried and mounted for counting since the phosphorus was carried in the precipitate as $FePO_4$.

The chemical treatment of the irradiated LiCl was identical with that for the sulfur with the exception that the first step, the nitric acid extraction, was omitted. The rest of the procedure of Kenny and Spragg was used, however, as well as the carrier separation described above.

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